FABRICATION & CHARACTERIZATION OF BIOMATERIALS FROM HYDROXYETHYL CELLULOSE SPONGES COATED WITH HYDROXYAPATITE

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Master of Science
(Advanced Materials)

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Universiti Malaysia PAHANG

Engineering • Technology • Creativity

SUPERVISOR'S DECLARATION

We hereby declare that we have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science (Advanced Materials).

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STUDENT'S DECLARATION

I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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ABSTRAK

Komposit *hydroxyapatite* telah diiktiraf sebagai bahan pengganti tulang dan gigi dalam bidang ortopedik dan pergigian kerana persamaan sifat kimia dan biologi untuk tisu keras manusia. Ia adalah bahan biokompatibel dan bioaktif yang boleh digunakan untuk memulihkan tisu *calcified* manusia yang rosak. *Hydroxyapatite* berliang mempamerkan ikatan yang kuat bagi tulang. Kewujudan liang menyediakan hubungan mekanikal yang membawa kepada penetapan firma bahan yang kukuh. *Hydroxyapatite* berliang adalah lebih resap dan lebih *osteoconductive* daripada bahan lain dan ia padat dan lebih baik kondisinya dalam bentuk berliang di atas kawasan permukaannya. Hal ini, amat membolehkan lebih banyak sel-sel untuk berhubung. Tesis ini memberi tumpuan kepada penghabluran dan pencirian hydroxyapatite (HA), pada span hydroxyethyl berliang (HEC). Span berliang HEC telah disediakan dengan kaedah beku-pengeringan. Span berliang (HEC) adalah polimer larut air yang tidak mengandungi bahan toksik dan biokompatibel. Hydroxyethyl selulosa (HEC) adalah gel seperti bahan yang digunakan secara meluas dalam industri. Penghabluran hydroxyapatite (HA) telah dilakukan dengan merendam span HEC ke dalam pelbagai kepekatan simulasi cecair badan (SBF) untuk tempoh masa yang berbeza. Untuk analisis bahan-bahan ini pelbagai teknik pukal dan alatan saintifik telah digunakan. Ia termasuk Mikroskop Imbasan Elektron (SEM), Fourier spektroskopi inframerah (FTIR), termogravimetri (TGA), dan mikroskop optik. FTIR telah digunakan untuk mengesahkan pemendapan apatite pada permukaan span HEC. HEC dengan kepekatan yang berbeza seperti 1% berat, 3% berat, 5 wt%, berat telah digunakan untuk membentuk span dan mereka disalut dengan HA, dan ini bertindak sebagai perancah. Hasil kajian telah mendapati bahawa keliangan, saiz liang dan liang antara sambungan bergantung kepada kepekatan HEC. Saiz kristal HA meningkat dengan kepekatan HEC. Span mempunyai liang dengan diameter ~ 2-60μm. Ujian invitro dengan sel pulpa gigi manusia (DPSC) juga dijalankan untuk menilai kesesuaian biologi perancah dan *scaffold*. Bioaktiviti menunjukkan bahawa perancah HEC/HA adalah bioaktif. Komposit selulosa hydroxyapatite-hydroxyethyl disintesiskan dengan proses pengeraman dalam garam akueus pada pH fisiologi dan suhu ambien. Menggabungkan hydroxyapatite ke hydroxyethyl selulosa boleh menghasilkan komposit dengan sifat-sifat mekanik dan kimia yang baik yang sesuai untuk pelbagai aplikasi-perubat.

ABSTRACT

Hydroxyapatite composites have been recognized as substitute materials for bone and teeth in orthopedic and dentistry field due to their chemical and biological similarity to human hard tissue. This biocompatible and bioactive material can be used to restore damaged human calcified tissue. Porous hydroxyapatite exhibits strong bonding to the bone, the pores provide a mechanical interlock leading to a firm fixation of the material. Porous hydroxyapatite is more resorbable and more osteoconductive than its dense counterpart and in porous form the surface area is greatly increased which allows more cells to be carried in comparison with dense hydroxyapatite. Most methods in producing HA used chemicals and some of them are very harmful to human being. This thesis is focused on the crystallization and characterization of hydroxyapatite (HA), on porous hydroxyethyl cellulose (HEC) which is a water-soluble, biocompatible and biodegradable polymer. Porous HEC sponges were prepared by freeze-drying method. Hydroxyethyl cellulose (HEC) is a gel like substance and is widely used in industries. The crystallization of hydroxyapatite (HA) was done by immersing HEC sponges into various concentration of simulated body fluid (SBF) for different time periods. For the analysis of these materials various bulk and particle level characterization techniques have been employed, which includes Scanning Electron Microscope (SEM), Fourier transform infrared spectroscopy (FTIR), Thermogravimetry (TGA), and mechanical testing. FTIR analysis were used to confirm the deposition of apatite on the surface of HEC sponges.HEC with different concentration like 1 wt%, 3 wt%, 5 wt%, were used to form the sponges and they were coated with the HA, and these acted as scaffolds. It was found that porosity, pore size and pore inter connectivity depends upon the concentration of the HEC. The size of the HA crystals increased with the concentration of the HEC. The sponges had pores with diameter~2-60μm and pores were interconnected. In-vitro testing with human dental pulps stem cell (DPSC) was also conducted to assess its biocompatibility. In-vitro bioactivity and biodegradability studies show that the HEC/HA scaffold was bioactive as well as bioresorbable. The hydroxyethyl cellulose-hydroxyapatite composite is synthesized by incubation in aqueous salt solutions at physiological pH and ambient temperature. Combining hydroxyapatite into hydroxyethyl cellulose may generate a composite with favorable mechanical and chemical properties that are appropriate for various medical applications.

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A	Cross-section area of specimens
d	spacing between atomic planes or lattice spacing (A^o)
E'	Storage modulus scaffolds (MPa)
E''	Loss modulus of scaffolds (MPa)
E^*	Complex modulus
tan δ	Loss factor or loss tangent
$\Delta E'$	Rigidity of polymers (%)
SR	Swelling ratio (%)
MR	Mass rate ratio (%)
Wh	Mass of sample after incubation in PBS
Wi	Mass of sample before incubation in PBS
Ws	Mass of swollen sample
Wd	Mass of dry sample

Greek Symbols

λ	X-ray wavelength
ε	Tensile strain
σ	Tensile stress
ω	Diffraction angle (degree)
σA	Sinusoidal stress
εA	Sinusoidal strain

LIST OF ABBREVIATIONS

CaP	Calcium phosphate
DPSC	Dental Pulps Stem Cell
DTG	Differential thermo-gravimetric
ECM	Extra-cellular matrix
EDX	Energy Dispersive X-rays
FESEM	Field emission scanning electron microscopy
FTIR	Fourier transforms infrared spectroscopy
GA	Glutaraldehyde
HEC	Hydroxyethyl cellulose
MTT assay	Colorimetric assay for cell metabolic activity
nHA	Nano-hydroxyapatite
PBS	Phosphate buffered saline
PVA	Poly (vinyl) alcohol
SBF	Simulated body fluid
SD	Standard deviations in the mean values
SEM	Scanning electron microscop
TGA	Thermo-gravimetric analysis
UTM	Universal testing machine
UV	Ultra violet light
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction

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